

Estimation of speciation and distribution of long-lived radionuclides in soils after irrigation with contaminated well water

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Abstract. In this study it is shown how the behaviour of radionuclides in agricultural soils can be quantitatively estimated. The sorption of those nuclides onto soil particle surfaces is a key process which every predictive model has to take into account. This has been accomplished by implementing a component additivity (CA-) model into the well known geochemical code PHREEQC that includes the most important sorbents clay, amorphous iron oxide and organic matter. For uranium and cesium it is shown that model calculations are in excellent agreement with experimental data.

1. INTRODUCTION

A current approach to estimate radiological doses and risks to humans living far in the future in the vicinity of nuclear waste disposals is the concept of so-called “Reference Biospheres”. Within this framework, the irrigation of agricultural soils by contaminated well water and the subsequent transfer of radionuclides within the biosphere is an important pathway to be considered.

The transfer of radionuclides into the food chain is strongly influenced by their speciation, distribution and transport within the soil. These processes depend on soil parameters such as pH value, mineralogical composition and organic matter. These parameters, in turn, may vary considerably with soil type and agricultural conditions. In this study it is shown that the distribution of radionuclides in soils after irrigation can be satisfactorily estimated by a CA (component additivity) model implemented into a geochemical code.

The concept of Reference Soils (e.g. the German Refesol system [1]) offers an attractive possibility of choosing representative soils that may be used to characterize a Reference Biosphere. By definition, Reference Soils are thoroughly specified and particularly their basic physicochemical properties are known. Our model is intended to be applied to these soils.

2. DESCRIPTION OF THE MODEL

In this work, modelling was done using the code PHREEQC [2], which has been frequently employed for geochemical calculations (see e.g. [3]). PHREEQC is also capable of calculating the transport of substances into the subsoil and can even be coupled to hydrological codes (e.g. Hydrus-1D for transport in the unsaturated zone [4]).

The distribution of radionuclides introduced into the soil is largely determined by sorptive soil components such as clay minerals, hydrous ferric oxides (HFO) and organic matter, the latter of which may be present in mobile as well as immobile form. Many components of the soil solution greatly influence sorption of radionuclides by competitive action; on the other hand they may increase solubility by complexation. An example is the phosphate ion (PO_4^{3-}) which is strongly sorbed to HFO surfaces [5]

and also complexes nuclides in solution such as uranium thus decreasing the respective distribution coefficient K_D .

In our model, illite has been chosen as a representative clay mineral which is frequently present in soils of temperate climates [6] and sorbs a variety of ions. The particular sorption constants were taken from the work of Bradbury and Baeyens [7–9] which considers exchange and surface complexation processes and also provides a model for the selective sorption of caesium on the frayed edge sites (FES) of illite.

Sorption on hydrous oxides was calculated using the well-known database of Dzombak and Morel [5], using the same surface complexation constants for sorption on ferrous and aluminium oxides.

Complexation by organic matter was taken into account by incorporating Model VI of Tipping [10], taking the constants for humic acid for the calculation of sorption by immobile organic matter and the constants for fulvic acid for the calculation of sorption by dissolved organic matter (DOM).

For the calculation of the number of the respective binding sites per kg soil water, the following parameters have to be provided:

- Cation exchange capacity (CEC)
- Oxalate extractable ferrous and aluminium oxides
- Organic matter content
- Clay content
- Soil density and porosity.

3. DETAILS OF THESE CALCULATIONS WILL BE SHOWN ELSEWHERE.

For the speciation and distribution calculation, the soil solution composition is also needed. The thermodynamic data base used here is an extension of the NAGRA database [11].

4. MODEL VERIFICATION

The model's suitability for the estimation of distribution of radionuclides in soil has to be verified. To this end, one would ideally need experimental data for all nuclides of interest obtained under controlled conditions. Obviously it is virtually impossible to find such a study that also supplies all necessary input data for simulation. However, Vandenhove et al. [12] studied the influence of soil parameters on soil solution concentration of one single nuclide, uranium, which is sorbed by clay, HFO and organic matter as well. They performed batch equilibrium experiments with 18 different soils that covered a comparatively wide range of soil parameters such as pH, CEC, clay content, HFO content and percentage of organic matter. Four weeks after contamination of these soils with ^{238}U , soil parameters were measured and the soil solutions were analysed. DOM was not measured; therefore, an average content of 36 mg/l (calculated from frequently found values given in Tab. 5.1-2 of the textbook by Scheffer and Schachtschabel [6]) was used for the PHREEQC simulations. Fe and Al content in solution was not given either, these parameters were estimated in the model by equilibrating the soil solution with Gibbsite and amorphous iron hydroxide.

Calculations were carried out by equilibrating the model system with the contaminated soil solution after equilibration with the uncontaminated solution. The major ion content of these solutions (including DOM) was assumed to be identical.

Fig. 1 shows how the PHREEQC simulations correspond to the data. The difference of the calculated U concentrations from the measured values lies within a factor of 2 (with the exception of one soil where the factor is 3). Considering the strong dependence of the U distribution in the soil solid/liquid system on various soil parameters [13], this result is remarkable.

A previous PHREEQC model was shown to satisfactorily predict caesium contents in the solutions of mineral soils [14]. The Bradbury and Baeyens model for illite [7] implemented here gave similar

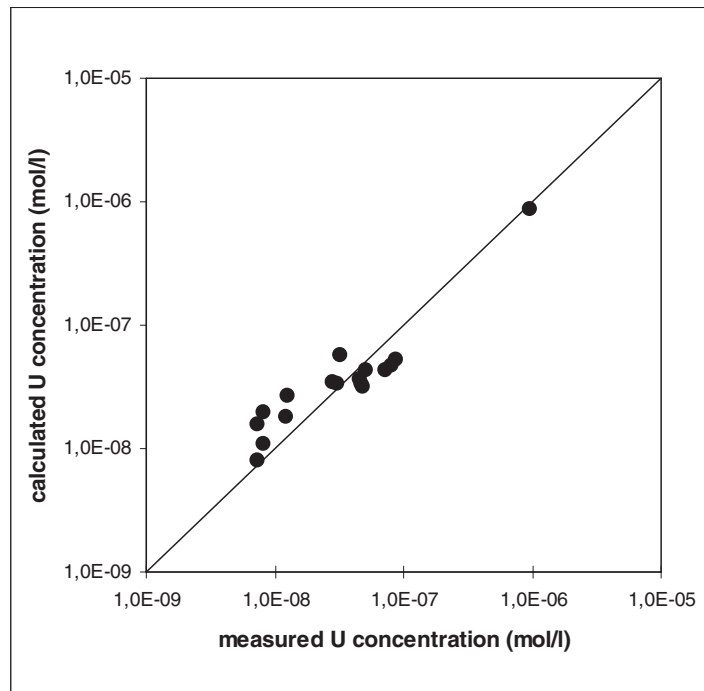


Figure 1. Comparison of measured and calculated U concentrations in soil solution.

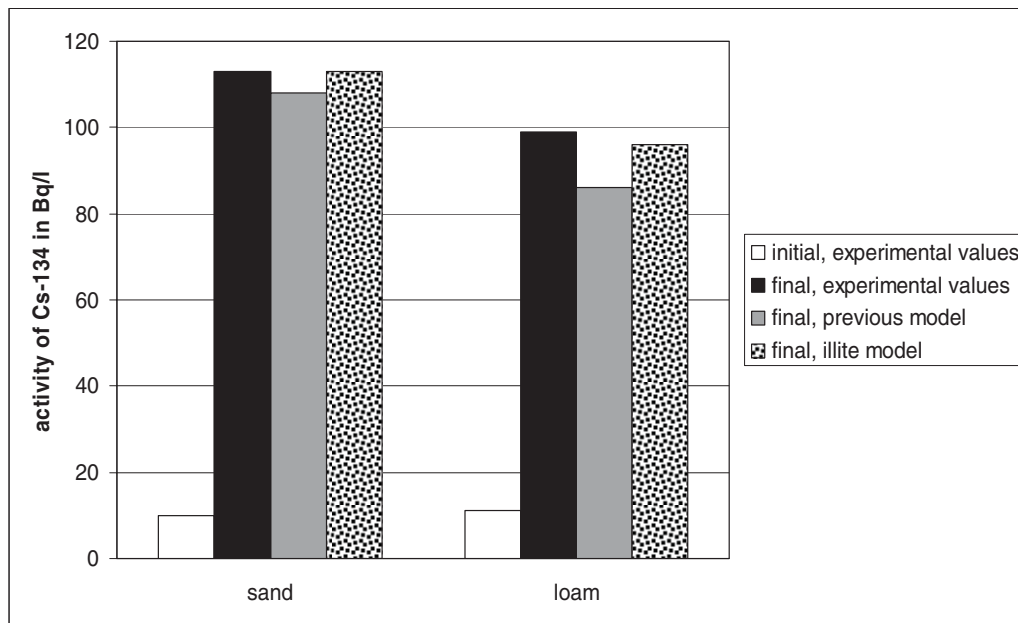


Figure 2. Activity of ¹³⁴Cs in soil solution before (white bars) and after application of 11.5 m potassium.

results (see Fig. 2). Details of the batch experiment shown in this figure and the previous model are given in [14].

5. CONCLUSIONS

The calculations presented in the previous section show that our model is capable of reasonably estimating the content of uranium and cesium in soil solution after contamination with these elements. This opens the possibility to predict the influence of soil parameters on the distribution of radionuclides in soils.

Acknowledgments

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